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# $PM_{2.5}$ source allocation in European cities: A SHERPA modelling study

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ABSTRACT

Many European cities suffer from poor air quality and still exceed the European standards prescribed by the Air Quality Directive, and the guidelines recommended by the World Health Organization (WHO). This is especially the case for $PM_{2.5}$, focus of this work. While international, national and local level actions to reduce air pollution have undoubtedly resulted in an overall improvement of the air quality over the years, there are still problems, which are localised in specific regions and many cities. A key issue is to determine at which scale to act in order to abate these remaining air pollution problems most effectively. Central to this, for cities, is a quantitative assessment of the different origins of air pollution (urban, regional, national and transboundary) to support the design of efficient, effective air quality plans, which are a legal obligation for countries and regions whenever exceedances occur.

The "Screening for High Emission Reduction Potentials for Air quality" tool (SHERPA) is used in this work to quantify the origins of air pollution in cities and regions, both from a spatial (urban, country…) and sectoral (transport, residential, agriculture…) perspectives. For $PM_{2.5}$ we conclude that (1) for many cities, local actions at the city scale are an effective means of improving air quality in that city; (2) the target sectors and scales to abate air pollution are city specific, even for cities that are located in the same country. Consequently, it is important to take into account these city-specific circumstances when designing air quality plans and (3) for many cities, sectoral measures addressing agriculture at country or EU scale would have a clear benefit on urban air quality.

## 1. Introduction

Many European cities suffer from poor air quality and regularly exceed both the European standards prescribed by the Air Quality Directive (EEA, 2017) and guidelines recommended by the World Health Organization (WHO). This is particularly the case for fine particulate matter ($PM_{10}$), for which both the daily (50 $\mu g/m^3$ not to be exceeded on more than 35 days a year) and the yearly average limit values (40 $\mu g/m^3$) are often exceeded in many cities and regions in Europe. For $PM_{2.5}$, the EU limit value (annual average of 25 $\mu g/m^3$) is generally met (EEA, 2017), but only few cities manage to keep concentrations below the levels recommended by the WHO (10 $\mu g/m^3$ on an annual basis). Adverse health effects and premature deaths are two of the major effects of poor air quality and current estimates suggest an average life loss of about 8–10 months in the most polluted European regions (e.g. Southern Poland, Po Valley, Benelux…) and cities. Consequently, air pollution remains the single largest environmental health risk in Europe (WHO, 2015).

Actions have been proposed and taken at the international (e.g. Amann et al., 2011) national (e.g. D'Elia et al., 2009) and urban scales (e.g. Giannouli et al., 2011) to reduce air pollution. While they have undoubtedly resulted in an overall improvement of the air quality over the years (Maas and Grennfelt, 2016), there are still problems which are localised in specific regions and cities (Amann, 2012). A key issue is thus to determine at which scale to act in order to abate these remaining air pollution problems most effectively. Central to this for cities, is a quantitative assessment of the different origins of air pollution in the city (urban, regional, national and international) to support the design of efficient and effective air quality plans, which are a legal obligation for countries and regions whenever exceedances occur.

Particulate matter ($PM_{2.5}$) is a pollutant that is either emitted directly (primary particulate matter) or formed through a series of complex chemical formation processes from other air pollutants (secondary particulate matter - see the review by Fuzzi et al., 2015 for more details). Depending on meteorological conditions, their residence time in the lower atmosphere is estimated to range from several days up to one

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week. It is worthwhile to note that for conditions other than the most stable low wind speed events, these residence times imply travel distances in the lower troposphere up to a thousand kilometres. In addition, air masses may follow complex trajectories. For example, under certain circumstances primary emissions from a city may react with precursors emitted elsewhere to form secondary particles that then return to the city where the precursors were emitted.

Chemistry transport models (CTM) can account for these complex transport, diffusion and chemical transformation processes and can therefore quantify the impact of cities on their pollution levels by performing simulations where emissions are switched on or off in a city. Because these models require intensive computational resources they are generally used to perform this detailed analysis for one city or region at a time. To cope with this limitation, the "Screening for High Emission Reduction Potentials for Air quality" tool (SHERPA) has been developed by the Joint Research Centre [Clappier et al., 2015, Thunis et al., 2016 and Pisoni et al. (2017)]. This simplified screening tool mimics a CTM, but with a much shorter time response. In this study, it is used to quantify the origins of air pollution in a large set of European cities. Both the spatial (urban, country…) and sectoral (transport, residential, agriculture…) contributions are quantified for 150 European urban areas, from where many of the current exceedances to the air quality EU and WHO limit values are reported. The focus is on urban background levels (meaning that road side traffic increments are not explicitly considered).

The work is structured as follows. Section 2 describes the methodology, in particular the SHERPA tool. In a second step, we analyse the spatial (Section 3) and sectoral source allocation for the 150 urban areas (Section 4). The main uncertainties associated to the source allocation results are presented in Section 5. Finally, in Section 6 we compare the results with those obtained in other studies. In particular, the differences with existing source apportionment work are discussed.

## 2. Methodology: the Sherpa modelling tool

SHERPA is a modelling tool, developed for the exploration of potential air quality improvements resulting from national/regional/local emission reduction measures. It is based on Source-Receptor Relationships (SRR). These SRR are a simplified version of a Chemistry Transport Model (CTM), used to simulate the contribution to concentration levels due to all precursor emissions ($NO_x$, NMVOC, PPM, $SO_2$ and $NH_3$) from one particular area of the domain (Clappier et al., 2015). They are used to estimate the effect of changes in precursor emissions on pollutant concentrations. In general, a SRR model consists of algebraic relationships between gridded precursor emissions and concentrations linked by a series of unknown coefficients that are identified based on a limited series of full CTM simulations (see details in Annex 1).

SHERPA assumes that the unknown parameters, describing the transfers between source and receptor cells, vary on a cell-by-cell basis but are not independent of each other. Instead the transfer coefficients are related through a bell shape function, assuming that the impact of emissions (within a source cell) on the concentration at a receptor cell decrease with distance between the source and receptor cells (Clappier et al., 2015; Pisoni et al., 2017).

Given its cell-to-cell characteristics, the SHERPA SRR formulation can be used to assess the impact of emission reductions over any given set of grid cells. Cities, regions or countries can therefore be freely defined in terms of boundaries, to test the effect of emission reduction policies on concentrations. At a given location, the contributions (primary or secondary, or sectoral) from the grid cell emissions belonging to the selected area (city, region or country) are then calculated explicitly. These contributions correspond to the impacts on air pollution that would result if emissions from a particular sector or spatial scale were reduced. It is important to note that while the final SRR simply connect emission to concentration changes, these SRR account for all processes modelled in the CTM (meteorology, chemical transformation…) that lead to the final concentration changes. In other words, SHERPA mimics the "dynamic" responses of a CTM for such emission reductions. We refer to this type of contributions as "source allocation", in contrast to 'mass-transfer source apportionment" contributions (referred in the following as "mass-transfer SA") that correspond to a "static" decomposition of the current air pollution levels. As shown by Clappier et al. (2017), the differences between the source allocation and mass-transfer SA approaches can be important when quantifying the role of precursors involved in chemical reactions leading to secondary particulate matter (see Section 6.1 for more details).

In its current configuration, SHERPA is based on the CHIMERE (Menut et al., 2014) model covering the whole of Europe at roughly 7 km spatial resolution. The anthropogenic emissions underlying the model simulations are based on GAINS total emissions per country-pollutant-sector for 2010, gridded with proxies from the MACC-TNO emission inventory (Kuenen et al., 2014) with residential sector emissions modified to account for the enhanced wood consumption at extremely low temperatures (Terrenoire et al., 2015). The meteorological input data is based on IFS (Integrated Forecasting System from ECMWF) for the year 2009.

Because of its simplifying assumptions and spatial resolution, SHERPA only calculates yearly average concentration levels of $PM_{2.5}$ for relatively large cities (covering a sufficient number of grid-cells). Additional information about the formulation of the SRR, their accuracy, their robustness and their validation process is available in Annex 1 and in Clappier et al. (2015), Thunis et al. (2016), Pisoni et al. (2017) and Thunis et al. (2018).

## 3. Spatial source allocation

It is of particular interest to know where urban pollution originates when designing air quality plans as it may help in choosing the right measures and the right spatial scale of implementation. To support this governance challenge, in this section we analyse the contribution from different spatial scales to city concentrations.

In terms of the spatial dimension, source allocation is analysed at three distinct scales: Europe (EU28 plus Switzerland and Norway, referred in the following as EUR30), the country and the "greater city" area. For the largest cities, an additional scale is considered with the split of the greater city into the "core city" and the peripheral "commuting zone".

Core cities are the local administrative units, with a population density above 1500/km$^2$ and a population above 50,000, where the majority of the population lives in an urban centre (OECD, 2012). Greater cities correspond to the functional urban areas (OECD, 2012) and consist of the core city plus the wider commuting zone, defined as the surrounding travel-to-work areas where at least 15% of the employed residents work in the city. Note that the commuting zone is limited in its extension to the country in which the city is located. It does not include transboundary commuting.

Given the SHERPA spatial resolution (~7 x ~7 km$^2$), only the largest city cores can be considered in the analysis. These include all EU28 capitals and other major urban areas within each country. Fig. 1 shows the final set of selected urban areas.

The SHERPA results are obtained in terms of the core city, greater city and country contributions (Fig. 2). Contributions are expressed in relative terms as a percentage of the urban concentration, taken at the city location where the highest modelled concentration level is found in that city. This choice is motivated by the fact that exceedances, and consequently air quality plans, must be designed considering the highest values within each air quality zone, according to EU Directive 2008/50/EC. Focusing on locations with the highest concentrations therefore helps targeting air quality plans where they are the most needed. Moreover, averaging source allocation results spatially over a city area might be misleading, because of the aggregation into a single

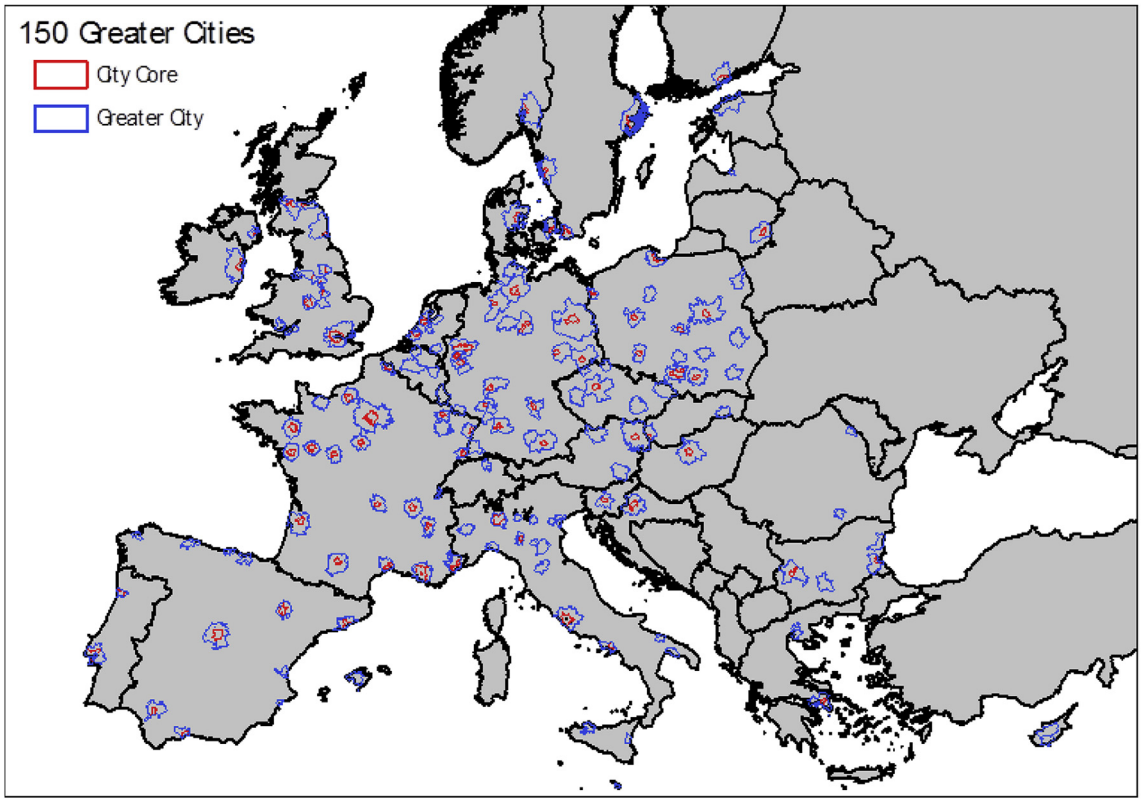


**Fig. 1.** Overview of the core city (in red) and greater city areas (in blue) for all 150 analysed cities. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

average number of very different allocations occurring across a city (this is emphasised for larger cities). Indeed, while source allocation at the city-centre will indicate a larger urban contribution than at locations at the city periphery, important differences are also likely to occur sector wise (e.g. increased traffic contribution at the city centre).

The key points that arise from this analysis are:

- The city core's contribution to annual $PM_{2.5}$ concentrations (at its location with the highest concentration) is on average (over the 84 considered cities) around 26%. The largest contributions are found in Milan (57%), Paris (56%), Madrid (52%), Mannheim-Luwigshafen (49%) and Warsaw (48%) and the lowest contributions in Burgas (6%), Leipzig (9%), Dresden (9%), Montpellier (11%) and Dusseldorf (11%). In general, local emissions are a significant contributor to annual PM concentrations in the largest EU cities, stressing the importance of air quality planning at the local level.
- At the greater city scale (city core plus commuting zone), cities' contribution to annual $PM_{2.5}$ concentrations at the most polluted spot is on average (over all 150 cities) around 31%. The largest contributions at that scale are found in Paris (66%), Madrid (65%), Athens (65%), Turin (63%) and Milan (63%), and the lowest in Nicosia (6%), The Hague (7%), Alicante (7%), Limassol (8%) and Heidelberg (9%). Therefore, actions taken at the greater city scale have a lot of potential in many cities. When the greater city area is considered, about 25% of the 150 cities contribute to at least 39% of their pollution while about 50% of the 150 cities contribute to more than 29% (Fig. 3).
- The contribution from the entire country (including city core and commuting zone) to annual $PM_{2.5}$ concentrations at the worst spot in the city is on average (over all 150 cities) around 56%. The largest contributions are found in Milan (89%), Warsaw (85%), Brescia (85%), Paris (84%) and Turin (82%) and the lowest in Valletta (12%), Limassol (14%), Nicosia (16%), Geneva (17%) and Palma de Mallorca (24%).

The remaining sources of emissions in this spatial analysis (transboundary, international shipping and/or natural) play a significant role in cities that are, either close to internal country borders, close to the EUR30 borders or under the influence of Saharan dust events. The largest contributions are found in southern European cities, subject to episodic dust events: Valletta (88%), Limassol (86%), Nicosia (84%) and Palma del Mallorca (76%) or in Geneva (83%) which, due to its geographic location, is largely affected by transboundary pollution.

## 4. Sectoral source allocation

Pollutant emissions originate from different human activities (like residential heating, transport, etc.) as well as from natural sources (e.g. dust, sea-salt, fires, etc.), as highlighted before. The sectoral allocation of $PM_{2.5}$ reported here distinguishes and quantifies the contributions from anthropogenic activity sectors and from natural sources as follows:

- Residential: this sector includes emissions from combustion in fireplaces, medium and single-house boilers, cooking and heating stoves in commercial, institutional and residential activities.
- Transport (road): this sector includes exhaust and evaporative emissions from light and heavy-duty vehicles and motorcycles as well as non-exhaust PM emissions due to road abrasion of tyres and brake wear.
- Agriculture: this sector includes emissions from livestock, fertiliser use and agricultural waste burning.
- Industry: this sector combines emissions related to combustion in energy industries (including public power, cogeneration and district heating), industrial combustion and industrial processes.
- Natural: This sector includes dust and sea salt.
- Others: The remaining emissions are grouped in a single category.

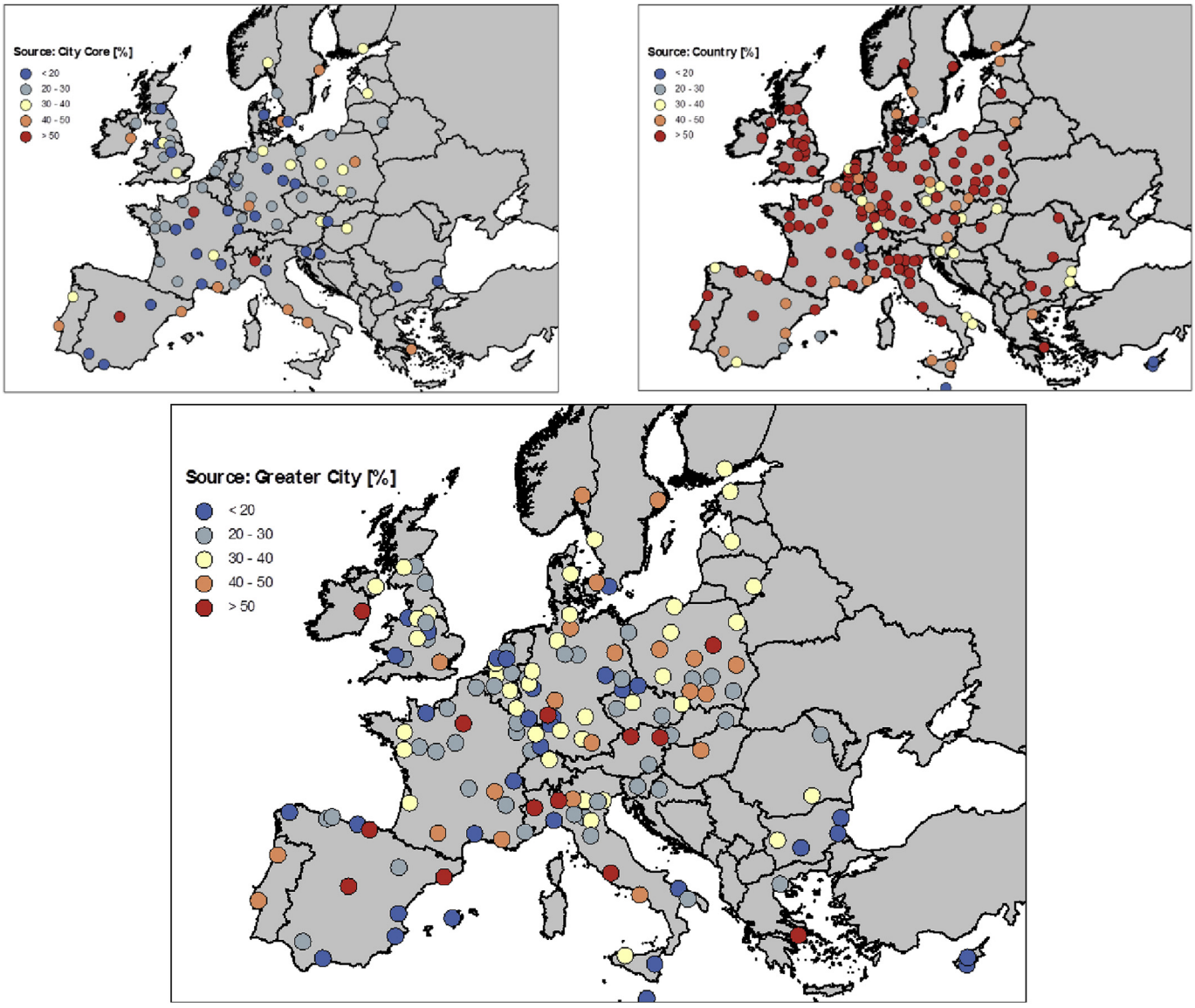


**Fig. 2.** Top-left: Contribution of city core emissions to urban $PM_{2.5}$ concentration (the 84 dots represent the urban areas where the city core is analysed); Bottom: Contribution of greater city emissions to urban $PM_{2.5}$ concentration (city core plus commuting zone) and top-right: Contribution of the country emissions to urban $PM_{2.5}$ concentration.

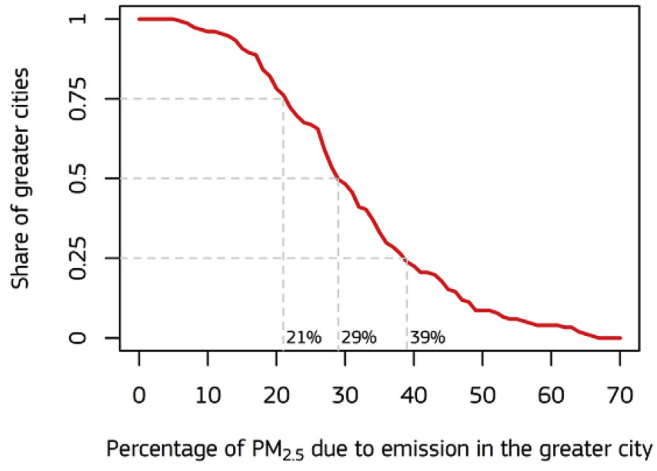


**Fig. 3.** Frequency cumulative distribution showing the share of greater cities (vertical axis) contributing to a given percentage of the $PM_{2.5}$ urban concentration. The 25, 50 and 75% percentiles are highlighted.

They include activities such as extraction and distribution of fossil fuels, solvent use, other mobile sources, machinery and waste treatment and disposal.

- External: This sector includes the international shipping as well as all emissions occurring outside the EUR30 domain.

We analyse here the relative contribution of these sectors to the $PM_{2.5}$ modelled urban background concentration levels reached in cities ( Fig. 4). The sectoral contributions are intended here as the overall impact of the emissions from a given sector, regardless of where these emissions originate (no spatial breakdown). For each urban area, results are presented at the location where the maximum concentration is reached within the city-core.

The key points arising from this analysis are synthesised below:

- The average contribution from the residential sector (top-left) in the 150 urban areas is 13%. The largest contributions are all found in Poland with the highest values in Warsaw (48%), Krakow (40%), Katowice (40%), Lodz (33%) and Poznan (33%). In general, the impact of residential heating is more important in the eastern

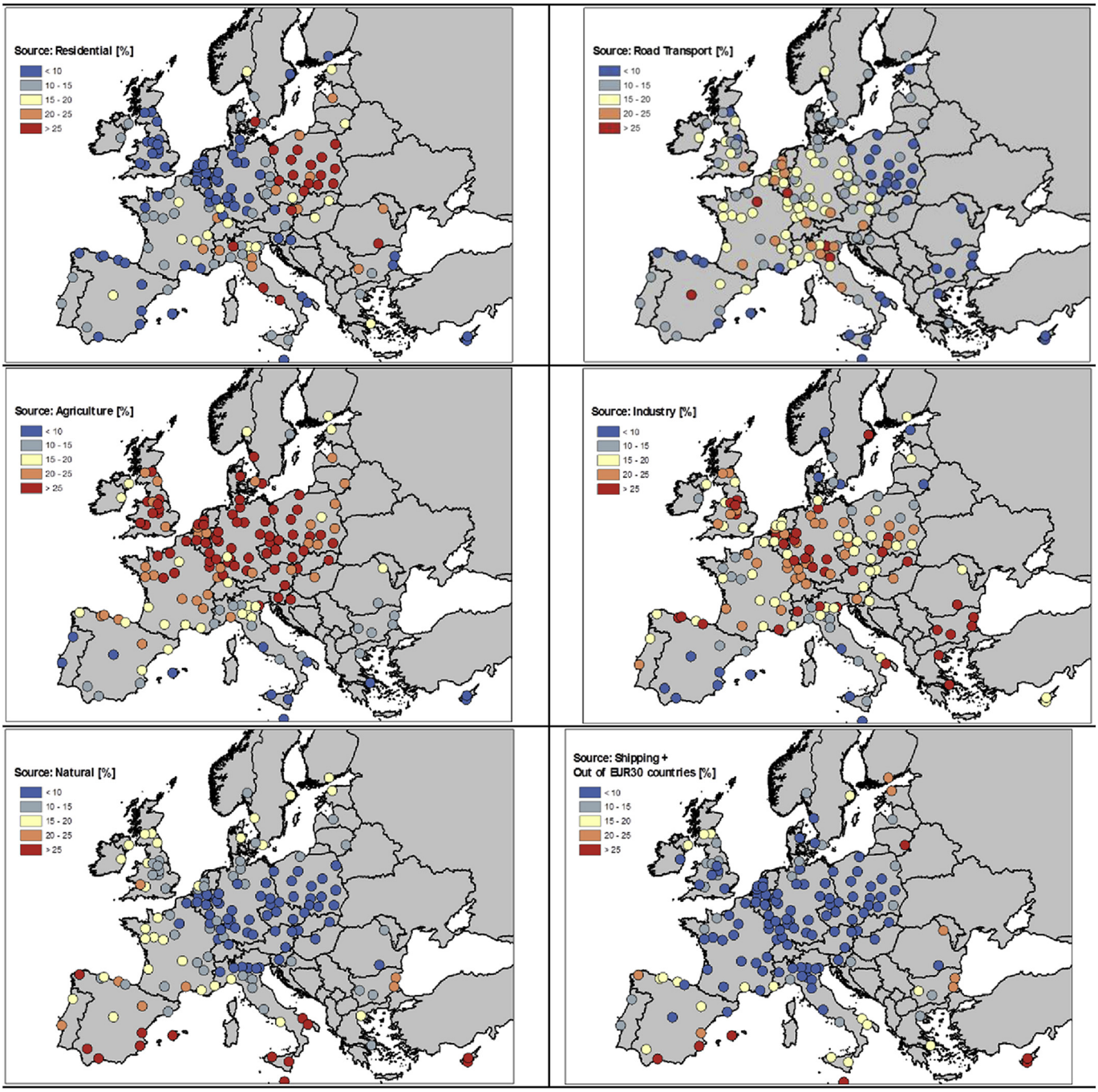


**Fig. 4.** Contribution of the individual sectors to the $PM_{2.5}$ urban background concentration. Top-left: residential, top-right: road transport, middle-left: agriculture, middle-right: industry, bottom-left: natural and bottom-right: remaining sources (shipping and outside EUR30). Each dot represents one urban area.

countries (Poland in particular) and in some cities in Italy. Northern countries (e.g. Scandinavia, UK, Belgium and Germany) do not show a large contribution from this sector.

- The average contribution from road transport (top-right) in the 150 urban areas is 14%. The largest contributions are found in Madrid (39%), Luxembourg (30%), Paris (29%), Verona (27%) and Bologna (27%). As expected, transport emissions represent an important contribution in some of the largest cities (Paris, Madrid, London). However, they also play a key role in densely populated areas like Belgium and the Netherlands. It is important to note that the numbers concern the transport contribution to urban background concentrations. At traffic stations (not analysed here), the contribution is likely to be proportionally larger.
- The average contribution from agriculture (middle-left) in the 150 urban areas is 23%. The largest contributions are all found among Germany and Czech Republic cities: Dresden (40%), Braunschweig-Salzgitter-Wolfsburg (39%), Usti nad Labem (38%), Plzen (37%) and Leipzig (36%). Agricultural activities and related emissions do not normally take place within the urban boundaries, although the greater city areas may include some agricultural areas. Nevertheless, the sector is responsible for a large fraction of the $PM_{2.5}$ concentration (secondary pollution) in many EU cities, especially in central Europe. This finding is in agreement with other work investigating the impact of agricultural emissions in urban areas (Bessagnet et al., 2014; Bauer et al., 2016).
- The average contribution of industry (middle-right) in the 150

urban areas is 20%. The largest contributions are found in Mannheim-Ludwigshafen (47%), Bilbao (46%), Linz (44%), Marseille (41%) and Brescia (37%). Industry plays a key role mostly in some of the Eastern countries (Bulgaria, Romania and Greece) as well as in the western part of Germany. It has a lower importance in southern Europe, although it appears as the key contributor in certain cities like Marseille and Turin.

- The average contribution of natural sources (bottom-left) in the 150 urban areas is 19%. $PM_{2.5}$ concentration peaks in cities in the Mediterranean area are associated with episodic dust events. The largest contributions are found in Valletta (46%), Limassol (43%), Palma de Mallorca (40%), Nicosia (39%) and Alicante (36%). Because natural components are mostly non-reactive, a comparison between the modelled source allocation obtained here and mass-transfer SA methods based on measurements (receptor models) is meaningful. The sum of the $PM_{2.5}$ relative contributions deriving from sea salt and dust in 32 urban areas quantified with receptor models (Belis et al., 2013) is on average 6% points higher than the one estimated in this study. The largest differences are observed in Atlantic and Mediterranean cities, suggesting that in these areas the role of natural sources could be higher than the one reported in this study. On the other hand, a recent mass-transfer SA work on five Southern European cities (Diapouli et al., 2017) reports percentage estimations that are most of the time below those reported in this work. A discussion on the potential sources of differences between the SHERPA methodology used in this work and mass-transfer SA is proposed in Section 6.1
- Other sources of emissions (external, i.e. from outside the EUR30, international shipping; bottom-right) play an important role in many cities located at the edge of the EUR30 domain (e.g. Burgas, Vilnius) but also in harbour cities where international shipping is a key contributor (e.g. Limassol, Alicante, Malaga).

When combining information from the spatial source allocation with its sectoral dimension, city diversity becomes very important as illustrated in Fig. 5.

In this figure, cities are divided into two categories: the first group includes all cities that have a contribution to their $PM_{2.5}$ pollution larger than 30% at the greater city scale while the second group includes the remaining cities that have lower impacts on their $PM_{2.5}$ pollution. Large cities (e.g. Paris, Madrid, Berlin and London) show a large impact on their air pollution with an important contribution from the transport sector. Cities in Poland, the Baltic republics and Italy also have important local contributions, often dominated by the residential sector. In Germany, many cities have local contributions dominated by the industrial sector. For cities that mostly import their pollution, one of the main causes of air pollution results from the emissions from the agricultural sector but many cities on the Mediterranean or Atlantic shores are influenced by natural (dust and/or sea-salt) sources as indicated above. This variability in terms of sectoral impact, even within a single country, illustrates the scope for targeting air quality plans on a city-by-city basis.

## 5. Underlying uncertainties and limitations

It is important to distinguish the uncertainties ascribable to the SHERPA SRR approach from those deriving from its input data (CTM, emission inventory, meteorology…). In this section, these two sources of uncertainties/limitations are detailed.

### 5.1. Uncertainties related to model input

#### 5.1.1. The underlying chemistry transport model

The SHERPA results strongly depend on the CTM used to define its SRR. The different approaches adopted to represent meteorological and chemical processes indeed vary from one model to the other, leading to potential uncertainties. Although the CHIMERE CTM base case scenario, in a similar configuration, has been extensively validated against observations (Bessagnet et al., 2016), it is not possible to validate CTMs for model-responses to emission changes. This is the reason why CTMs are regularly tested in the frame of inter-comparison exercises (Cuvelier et al., 2007; Thunis et al., 2007; Vautard et al., 2007). This process of inter-comparison is continuously on-going, to increase the reliability and robustness of the whole approach.

#### 5.1.2. Spatial resolution

The CHIMERE simulations within SHERPA are run with a 7 km spatial resolution. Schaap et al. (2015) showed that this resolution was accurate enough to capture urban background concentrations, the focus of this work. This spatial resolution however limits the analysis to the largest EU cities as smaller cities might cover too few grid cells. This is why a threshold of about 300 $km^2$ (~6 grid cells) is applied on the city selection. Note that emissions within a grid-cell, crossed by the city (or greater city, country) boundaries, are attributed to the city proportionally to the city area included within the grid cell.

#### 5.1.3. Absolute vs. relative source allocation

One of the findings of past modelling inter-comparison exercises (e.g. CityDelta, EuroDelta, Cuvelier et al., 2007; Thunis et al., 2007) is that relative fractions (i.e. concentration change divided by concentration) are generally more robust than absolute values (concentration). This is because concentration changes and concentrations are generally correlated (an overestimation of the concentration is likely to lead to an overestimation of the concentration change as well). All results are therefore expressed in terms of relative fractions.

#### 5.1.4. Meteorological variability

The results presented in this article are based on a single meteorological year (2009). Although this year is thought to be representative of average meteorological conditions, the current set-up does not account for inter-annual variability. In this respect, repeating the analysis with other years would increase the robustness of the results. The fact that all results are presented in terms of relative fractions, however, reduces the possible impact of inter-annual variability.

#### 5.1.5. Emissions

The results presented here strongly depend on the quality of the underlying emission inventory. Uncertainties in emission inventories are known to be high, especially at the urban scale, as highlighted by Trombetti et al. (2018) who compared the features of different EU wide top-down inventories over major European urban areas. In this respect, we hope that the results presented here can be used to trigger the discussion with local air quality managers to detect possible inconsistencies and support the improvement and the harmonisation of the underlying emission inventory. Another source of uncertainty is related to the emission reference year (2010) which does not account for emission changes that occurred in the most recent years.

While uncertainties and limitations can rather easily be identified, their quantification remains challenging because additional simulations are required to study specific sensitivities (e.g. new simulations with other emission inventories, meteorology…). In this respect, work is in progress to assess and quantify the uncertainties that are related to the use of two different CTMs driven by different emissions and different meteorology's (publication in preparation).

### 5.2. Uncertainties related to the SHERPA approach

The main added value of SHERPA lies in its ability to mimic CTM responses to emission changes in few minutes of calculation instead of the few days usually required by a CTM simulation. These performances are obtained at the expense of two main assumptions:

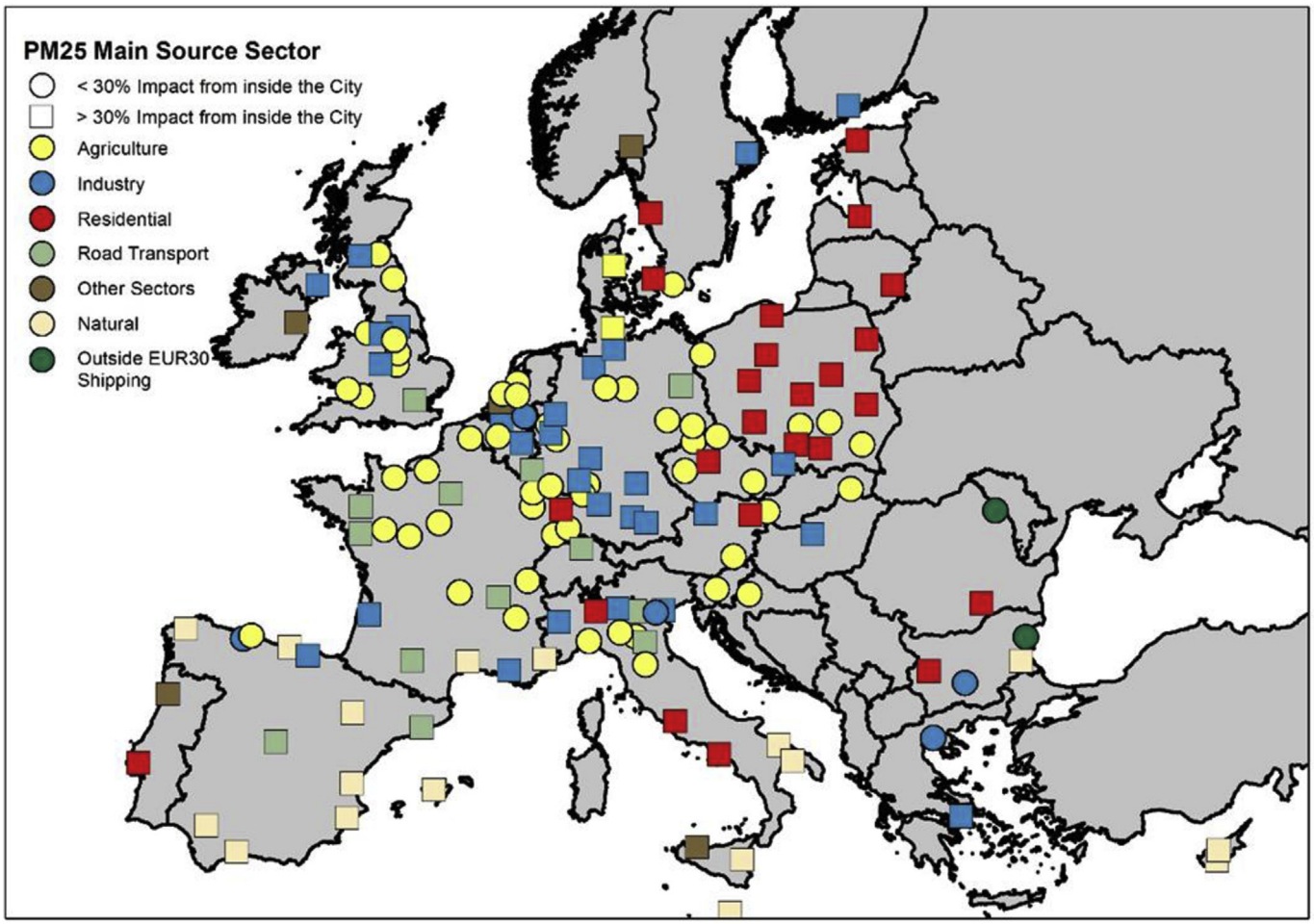


**Fig. 5.** Sectoral source allocation in 150 cities. Cities that have a contribution to their $PM_{2.5}$ pollution larger than 30% are indicated by squares while the remaining cities that have a lower impact on their $PM_{2.5}$ pollution are represented by circle. The dominant sector in terms of contribution is indicated via the color scheme. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

*5.2.1. The relationship between emission and concentration changes is linear*

Given the non-linear chemical processes leading to the formation of secondary PM, we expect non-linear relationships between PM concentrations and their precursor emissions. In their work, Thunis and Clappier (2015) have shown that these relationships can be assumed linear when concentrations are averaged over long periods (seasonal or annual averages). However, it is important to note, that these tests were performed for emission reductions not exceeding a 50% percent threshold. Beyond these levels, non-linearity might become more important, limiting the range of application of the approach. This issue is further discussed in the next Section.

*5.2.2. A simple "bell shape" function spatially links emission and concentration changes*

Statistical analysis shows that the correlations between PM concentrations and their precursor emissions decrease with distance following a "bell shape" function (Pisoni et al., 2017). This "bell shape" function is used to express the coefficients of the SRR as a function of the distance between sources and receptors. This simple "bell shape" function is precursor and grid cell specific.

These two assumptions have been assessed through a validation process, which covered test cases in many EU cities, regions and countries. This validation highlighted the good agreement between the SHERPA SRR and the full CHIMERE model (Pisoni et al., 2017).

In addition to these two main assumptions, the following two points must be made:

*5.2.3. International shipping and external sources*

Although the SHERPA modelling accounts for the impact of emissions from international shipping and from countries outside the EUR30 domain (boundary conditions), these contributions cannot be distinguished from each other. They are grouped in a category named "external". This external impact is important in harbour cities, where international shipping emission can be important and in urban areas that are close to the EUR30 borders.

*5.2.4. High-level sources and surface sources*

SHERPA distinguishes sectoral impacts but high-level sources (for which the release height becomes an important element) and surface sources are treated similarly. This might introduce some uncertainty in the sectoral breakdown when both surface- and high-level emission sources are present in a same sector.

With the use of a sensitivity approach, Pisoni et al. (2018) showed that the most influential inputs of the whole SHERPA modelling approach are by far the policy selection (i.e. the level of ambition to be considered in the design of the air quality plan, not considered in this work) and the emissions, in particular of PPM, $NO_x$, and $NH_3$. The SHERPA model coefficients ($\alpha$ and $\omega$, see Annex 1 for details) are the least influential inputs. This stresses the need of improving the quality of the underlying emission inventories but it also highlights the robustness of the SHERPA assumptions.

## 6. Comparison with other approaches

Two of the main findings of this study (detailed in Sections 3 and 4) are namely that (1) many EU cities have a major role to play at their own scale to abate air pollution, and that (2) agriculture is a key contributor to air pollution in many EU cities. These two findings sometimes contradict those from other studies. In this section, we show that most of the differences mainly result from the methodological approach rather than from the uncertainties and limitations detailed in the previous Section. We analyse, in particular, the differences between the source allocation results obtained in this work with those obtained in other studies on the basis of alternative methodologies.

First, differences are analysed with respect to methodologies based on mass-transfer SA, with a focus on the impact of agricultural emissions. In a second step, we explore the differences with methodologies based on the incremental approach, with a focus on the importance of the local contribution to air pollution in cities.

### 6.1. Comparison with "mass-transfer SA" approaches

As mentioned earlier, we use "source allocation" as the terminology to characterise the SHERPA results to distinguish them from the more usual "mass-transfer SA" approach. In this section, we discuss the main differences between the two approaches and Annex 2 describes a simple example to illustrate those differences.

Both mass-transferSA and source allocation aim at quantifying the contributions of different emission sources to the concentration of a given pollutant at one location.

Mass-transfer SA is computed using receptor models (e.g. Belis et al., 2013) or tagging-species algorithms built-in CTMs (e.g. Kranenburg et al., 2013). This approach decomposes the concentration into a sum of contributions, each associated to a given emission source. In the tagging species algorithms the decomposition is done by attributing the mass of each fraction composing the PM concentration (e.g. $NO_3$, $NH_4$) to its associated emission precursor (e.g. $NO_2$, $NH_3$), hence the attribute "mass-transfer" as it is designed to compute the contributions of the sources in terms of mass transferred from the source to the receptor. By construction, mass-transfer SA apportions the entire mass of the pollutant to its sources and therefore lead to contributions that depict a 100% effect of the sources. Clappier et al. (2017) showed that this was equivalent to concentration changes resulting from a 100% change in emissions from all sources, simultaneously. Consequently, mass-transfer SA contributions do not provide information on how the concentration would change for any other levels of emission reductions and/or for different combination of sources (see Annex 2 for details).

Source allocation also decomposes the effect of a simultaneous reduction of all precursors into a sum of precursor contributions. However, each precursor contribution is computed with a CTM by reducing each precursor emissions independently by a given percentage, which must be low enough to guarantee a linear relationship between emissions and concentrations changes (see Annex 2 for details).

When the relationship between emission and concentration is linear through the entire range of reductions (from 0 to 100%), mass-transfer SA and source allocation provide similar results whatever the percentage of reduction used for source allocation (Clappier et al., 2017). Conversely, as shown in the Annex 2, mass-transfer SA and source allocation results differ when non-linearity occur (e.g. for secondary PM formation). Under those non-linear conditions (e.g. secondary PM), the Authors concluded that mass-transfer SA was not suited to the purpose of air quality planning while source allocation was suited to this purpose as long as the reduction percentage was kept low enough to guarantee linearity between emission and concentration.

In the case of SHERPA, the source receptor relationships are based on full CTM scenarios performed with emission reductions around 50%. Up to this level of reduction, non-linearities remain below 10% (Thunis and Clappier, 2015). The SHERPA results are therefore valid for this range of emission reductions and provide information on how air quality would change when emission abatement actions are applied. Such information is of direct relevance to air quality planning.

Finally, it is important to note that the differences between the two approaches in our example (see Annex 2) are important because of its focus on secondary particulate matter reactions that only involves non-linear processes. We must keep in mind, however, that the fraction of secondary PM compounds in the atmosphere is large and sometimes dominates the primary fraction. These cases are therefore not exceptional. One of the consequences of these differences is that the SHERPA results (but also other source allocation results) lead to agricultural contributions to $PM_{2.5}$ that are much larger than those resulting from mass-transfer SA. For instance, in the city of Berlin, SHERPA indicates values close to 30% while mass-transfer SA estimates it below 5% (Berlin, 2014).

### 6.2. Comparison with the "incremental approach"

The incremental approach consists in assuming that the urban impact of a city on its own air pollution can be well estimated by the difference between the concentrations at two background locations (city and rural). This type of approach is widely used (e.g. Petetin et al., 2014; Kiesewetter et al., 2015, Berlin, 2014). Thunis (2018) recently showed that the incremental approach accurately represents the urban impact only if two assumptions are fulfilled: (1) The city emissions have no influence on the rural location, and (2) the background (concentration obtained when the city emissions are switched off) is identical at the rural and city locations. Assessing the validity of these two assumptions for a series of cities, Thunis (2018) showed that the incremental approach leads to an underestimation of the local contribution ranging from 30 to 50% for medium and large size cities. SHERPA therefore delivers urban contributions that are larger for many cities than the estimates reported by incremental-based methods. In the case of the GAINS results (Kiesewetter et al., 2015), it is worth noting that the assumption attributing the secondary PM fraction entirely to a regional origin leads by construction to the non-fulfilment of the first incremental assumption mentioned above, as it implies that the secondary gas-phase PM precursors ($NO_x$, $SO_2$, …) emitted by the city influence the rural location. The same assumption also results in different shares being allocated to the urban sectors, as compared to SHERPA. The importance of this secondary PM fraction is obviously city-specific but can be important as highlighted by Lenschow et al. (2001) who estimated the local production of secondary inorganic aerosols (SIA) to 5.6 $\mu g/m^3$, i.e. 43% of the total urban background SIA. As a result, the SHERPA sectoral allocation will give less importance to urban sectors characterised by a high share of PPM emissions (e.g. wood burning) and more to urban sectors emitting gas-phase precursors (e.g. transport).

These considerations are further detailed and discussed in Thunis et al. (2018).

## 7. Conclusions

There is a need to provide information to improve air quality policy governance, to support authorities in choosing the most efficient actions at the appropriate administrative level and scale. In particular, an appropriate balance between local actions focusing on the urban scale and actions requiring national/international efforts need to be found. The purpose of this work is to provide, based on the SHERPA tool (Thunis et al., 2016), city specific source allocation information on annual $PM_{2.5}$ concentrations in terms of sector and spatial dimensions for 150 cities in the EU. The main findings are:

- *For many cities, local actions at the city scale are an effective means of improving air quality in that city*. Almost half of the considered cities

(73 out of 150) have the potential to reduce their annual PM concentration by 30% or more through local (greater city) actions. In fact, there is a significant number of cities where the share of their own contributions to PM pollution is even higher: ≥ 40% impact (34 cities) and ≥50% impact (13 cities). Long-range transport is important, particularly in cities located near the EUR30 boundaries. However, the overall finding is that cities have an important role to play by taking actions at their own scale. It is important to emphasise that the emissions in cities contribute significantly to country and EU overall PM background concentrations, reinforcing the important role of cities in reducing the air pollution through a multilevel approach.

- *Target or key sectors and scales to abate air pollution are city specific.* Cities differ in the way in which their PM concentrations respond to abatement measures, even when located in the same country. Actions taken at different scales or in different activity sectors lead to impacts that depend on the city. Given that measures have so varied effects in different cities, there is a clear need to take into account these city-specificities when designing air quality plans. Actions that are efficient in one city might not be efficient in others.
- For many cities, sectoral measures addressing agriculture at country or EU scale would have a clear benefit on urban air quality. Although agricultural activities take place mostly outside the "city" boundaries, as defined here, agriculture emissions considerably impact air quality in many EU cities. Agriculture contributes to more than 30% of the air pollution ($PM_{2.5}$ concentrations) in about 21% of the cities (31 cities out of 150) and to more than 20% of the pollution in 66% of the cities. The extent of the impact of agriculture on air quality is indicative of the potential of EU- and/or country-wide measures addressing this sector.

Moreover, other sectoral measures can have an important potential at the urban scale even though they are applied at EU or country scale. This is the case i.e. of road transport where the EURO norms are, in practice, most effective in the areas where traffic is most important, i.e. cities.

About half of the reported EU exceedances occur in the areas covered by the 150 greater cities included in this study. However, many smaller urban areas are not considered in this work. It is therefore important to extend this work in the future to these smaller urban areas as they may have their own specific characteristics. Similarly, within cities, a proper estimate of the contributions at traffic stations would complete this analysis at the local scale.

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## Annexes

### *SHERPA formulation and validation*

*Formulation*

For a fast time-response, integrated assessment tools are based on Source-Receptor Relationships (SRR). SRR are a simplified version of a CTM, used to give the contribution to concentration levels of a particular pollutant from precursor emissions from one particular area of the domain. They are used to estimate the effect of changes in precursor emissions on pollutant concentrations. In general, a SRR model consists of algebraic relationships between gridded precursor emissions and concentrations.

The most precise way to use a CTM to produce SRR for the model domain would be with an independent grid cell-to-grid cell approach. This involves simulating independently the effect of emissions changes in each single grid cell and would require changing precursor emissions in individual grid cells one at a time and looking at the resulting change in concentrations in each receptor cell.

Formally, a concentration change in receptor cell “j” is the sum of the concentration changes due to changes in precursor emissions “p” emitted from any source cell “i” within the domain, so that the concentration delta in a receptor cell “j” can be computed as follows:

$$\Delta PM_j = \sum_{p}^{N_{prec}} \sum_{i}^{N_{grid}} a_{ij}^{p} \Delta E_i^{p}$$

where $N_{grid}$ is the number of grid cells within the domain, $N_{prec}$ is the number of precursors, $\Delta E_i^p$ and $\Delta PM_j$ are the emission and concentration deltas, and $a_{ij}^p$ are the unknown parameters to be identified that represent the transfer coefficients between each source cell $i$ and receptor cell $j$.

While theoretically very simple, the resulting number of unknown parameters ($a_{ij}^p$), describing the transfers between source and receptor cells that need to be identified is very large and would require a very costly series of independent CTM runs to solve the system.

In SHERPA, it is assumed that the unknown parameters (i.e. the $a_{ij}^p$ coefficients) vary on a cell-by-cell basis but are not independent of each other. The $a_{ij}^p$ coefficients are assumed to be related through a bell-shaped function that accounts for variation in terms of distance but is directionally isotropic:

$$a_{ij}^{p} = \alpha_j^{p} (1 + d_{ij})^{-\omega_j^{p}} \quad (2)$$

where $d_{ij}$ is the distance between a receptor cell “$j$” and a source cell “$i$”.

The choice of the bell-shaped function is supported by an analysis of the correlation between emission changes and concentration changes (Pisoni et al., 2017). However, this assumption is only reasonable for yearly average emissions and concentrations and can be explained by the fact that less frequent low wind speed – high concentration episodes, which are inherently more isotropic, contribute proportionately much more to the annual mean concentration than the more frequent high wind speed – low concentration episodes.

Thus, in SHERPA the matrix of transfer coefficients from source cells to a receptor cell is replaced by a simple function with two parameters for each precursor and all coefficients are known when the two parameters $\alpha$ and $\omega$ are identified for a given receptor cell $j$ and a given precursor $p$. These two parameters $\alpha$ and $\omega$ are the amplitude and width of the function respectively. $\alpha$ can be interpreted as the relative importance of each precursor $p$ in producing the pollutant concentration in cell $j$, whereas $\omega$ captures how the contribution of the precursor $p$ emissions decreases with distance from cell $j$. Because the bell-shaped function only depends on distance $d_{ij}$, all cells that are at a particular distance from the receptor have identical contributions per unit emission to the annual average concentration of the receptor cell. The bell shape function however differs for each receptor grid cell and emission precursor. The final formulation implemented in SHERPA is:

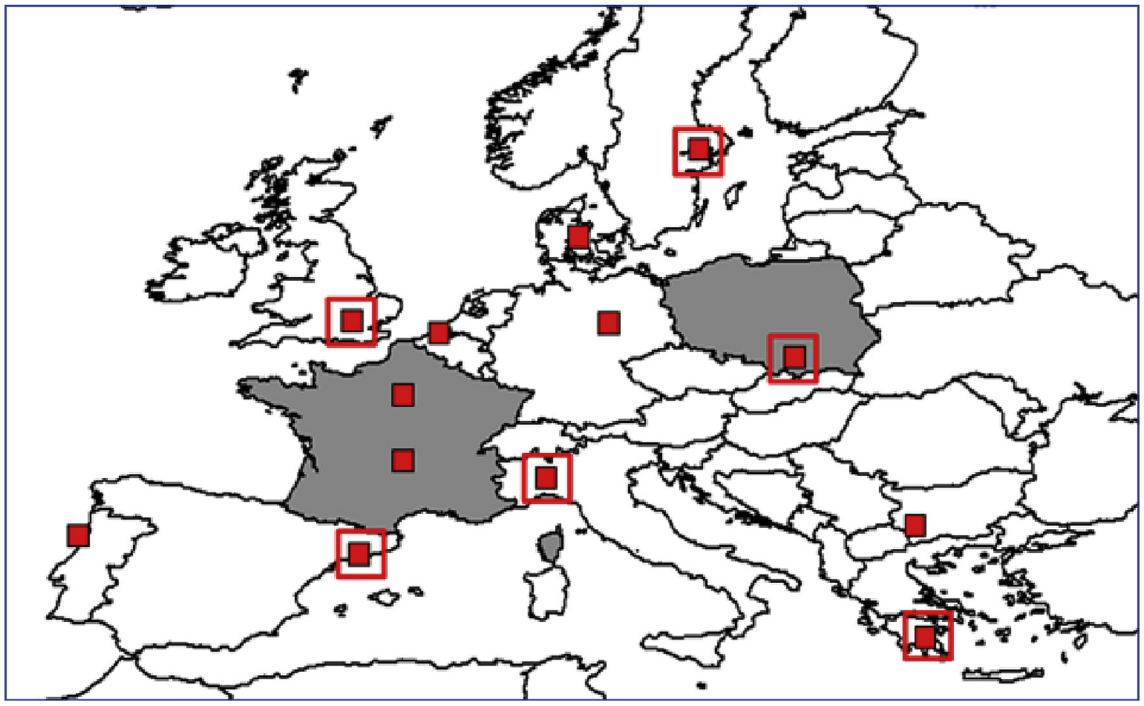

**Fig. 6.** Areas where emission reduction where applied in specific CTM simulations for validation purpose (countries in grey, regional areas (large squares) and local areas (small squares).

$$\Delta PM_j = \sum_{p}^{N_{prec}} \sum_{i}^{N_{grid}} \alpha_j^p (1 + d_{ij})^{-\omega_j^p} \Delta E_i^p$$

With the SHERPA approach, the number of unknown parameters is equal to 2 ($\alpha$, $\omega$) for each precursor and receptor cell "j". Consequently, for the five precursors of $PM_{2.5}$ (VOC, $SO_2$, $NO_x$, PPM and $NH_3$), ten independent CTM simulations are needed for a given receptor cell, provided that they deliver independent information. The same CTM scenarios can be used to identify $\alpha$ and $\omega$ for all cells within the domain.

Currently, the CTM scenarios are performed at 7 km resolution by reducing emissions by a given percentage (β) over the entire European domain with each precursor being reduced independently or in combination. In order to guarantee independence throughout the domain, so that the solutions are valid for all grid cells, 19 runs are used rather than the theoretical minimum of 10. It is further assumed that linearity applies so that the reductions initially obtained for a reduction $\beta$ can be extrapolated to any other level of reduction below that threshold.

Given its cell-to-cell characteristics, the SHERPA formulation can be used to assess the impact of emission reductions over any given set of 7 km grid cells. This flexibility in terms of geographical areas is a direct consequence of the cell-to-cell SRR.

*Validation*

The only way to ensure that the SHERPA assumptions described above are acceptable in practice is to perform a thorough evaluation process. It is important to note that the main questions addressed here: 1) which is the fraction of pollution originating from urban areas? or 2) which is the fraction of pollution originating from a given sector? cannot be answered by means of air quality measurements. As with other simplified SRR models with similar scope [e.g. GAINS (Amann et al., 2011), RIAT (Carnevale et al., 2012)], the only possible evaluation consists therefore in comparing the simplified model (SHERPA) with the full CTM for a series of targeted emission reduction scenarios. For SHERPA, these scenarios have been selected to cover a wide range of situations with emissions reduced in different countries, regions and cities (Fig. 6).

The validation simulations were performed for each precursor ($NO_x$, PM …) to ensure that the different CHIMERE model components (e.g. primary and secondary) are reproduced accurately. In total, 84 different cases [(2 countries + 6 regions + 13 cities) *4 precursors] are used for validation.

The usual validation is made by comparing the concentration levels for a given emission scenario that is independent from the CTM simulations used to derive the simplified SRR. Such a comparison is shown in Fig. 7, where the agreement obtained between SHERPA and the full CHIMERE results is shown in terms of concentration deltas for different independent scenarios. These scenarios cover different spatial scales: country, regional and local, different precursors and different sectors. Only a sample of these validations is shown here but additional examples may be found in Thunis et al. (2018).

*Mass-transfer SA and source allocation*

In their publication, Clappier et al. (2017) highlight the main differences between two methodologies: sensitivity analysis and mass-transfer SA. We propose to extend this comparison to source allocation with a simple theoretical example as in Clappier et al. (2017). This example focuses on the formation of secondary particulate matter (PM) in the atmosphere, limited to one reaction between nitrogen oxides ($NO_2$) and ammonia ($NH_3$):

$$NO_2 + NH_3 \rightarrow PM[NH_4NO_3]$$

From this reaction, 1 mole of PM is produced by the combination of 1 mole of $NH_3$ and 1 mole of $NO_2$. We also limit our example to emissions

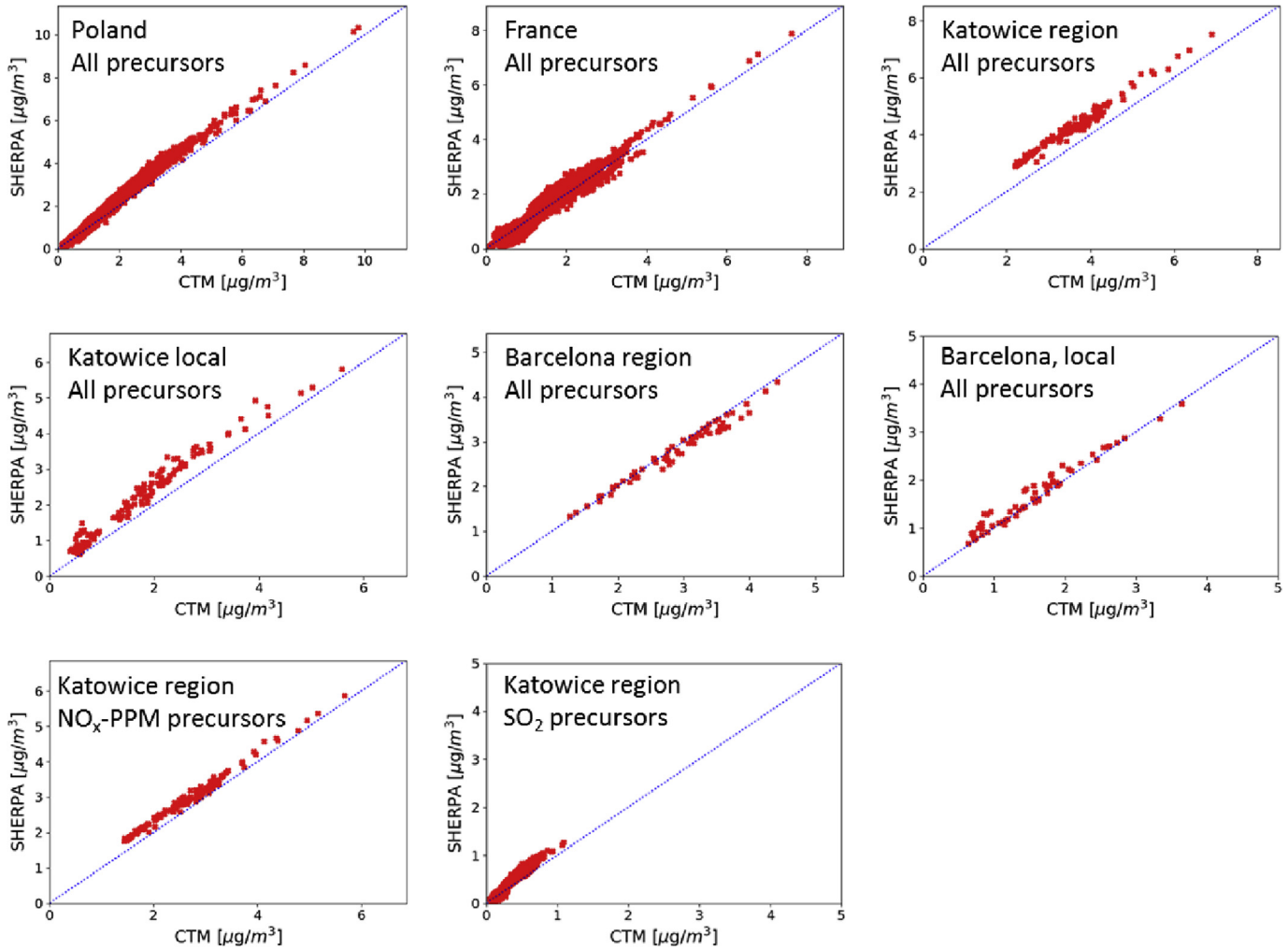


**Fig. 7.** Validation scatter plots for SHERPA for different countries (France and Poland), regional and local domains around the cities of Barcelona and Katowice. Results are shown in terms of concentration changes against those obtained with the full CTM CHIMERE model. Emission reductions include either the whole set of precursors (first two top rows) or focus on specific precursors ($NO_x$ & PPM or $SO_2$). It is important to stress that these validation scenarios are independent from the scenarios used to generate the simplified SRR. Values are expressed in terms of concentration in μg/m$^3$, comparing the CTM (x-axis) and the SHERPA SRR (y-axis). Each dot represents a grid cell value.

from two activity sectors: the transport sector (T) assumed to only emit $NO_2$ and the agricultural sector (A) assumed to only emit $NH_3$. We further assume (for convenience) that no background pollution is present (i.e. there is no PM when all emissions are zero). The initial conditions correspond to an availability of 100 moles for $NO_2$ and 50 moles for $NH_3$ leading to the formation of 50 moles of $PM[NO_3NH_4]$, but the reasoning remains valid for other initial conditions.

**Sensitivity analysis** is here intended as the methodology proposed by Stein and Alpert (1993) that separates the impact of a simultaneous reductions of different precursors emissions into precursor-related and interactions components. Emission reductions can address a specific precursor (like $NO_2$ or $NH_3$) or activity sector (like transport or agriculture) or be limited to a specific area (like a city, a region or a country). In our simple example, the $PM[NO_3NH_4]$ results only from two precursors ($NO_2$ emitted by transport and $NH_3$ emitted by agriculture), leading to the following decomposition:

$$\Delta M^{T\&A(\alpha)}_{NH_4NO_3} = \Delta M^{T(\alpha)}_{NH_4NO_3} + \Delta M^{A(\alpha)}_{NH_4NO_3} + \hat{M}^{int(\alpha)}_{NH_4NO_3}$$

where $\Delta M^{T\&A(\alpha)}_{NH_4NO_3}$ is the reduction in $PM[NH_4NO_3]$ obtained when the transport ($NO_2$) and agriculture ($NH_3$) emissions are reduced simultaneously by a percentage α while $\Delta M^{T(\alpha)}_{NH_4NO_3}$ (resp. $\Delta M^{A(\alpha)}_{NH_4NO_3}$) is the reduction in $PM[NH_4NO_3]$ obtained when only the transport (resp. agriculture) emissions are reduced by a percentage α. Due to chemical interactions between $NO_2$ and $NH_3$ the impact of a simultaneous reduction of these two precursors is generally not equal to the sum of the impacts of each precursor individual reduction ($\Delta M^{T\&A(\alpha)}_{NH_4NO_3} \neq \Delta M^{T(\alpha)}_{NH_4NO_3} + \Delta M^{A(\alpha)}_{NH_4NO_3}$). The interaction term is computed as follows:

$$\hat{M}^{int(\alpha)}_{NH_4NO_3} = \Delta M^{T\&A(\alpha)}_{NH_4NO_3} - \Delta M^{T(\alpha)}_{NH_4NO_3} - \Delta M^{A(\alpha)}_{NH_4NO_3}$$

Let's calculate the different terms for two levels of reduction (α):

- If α = 20%, the transport ($NO_2$) emissions decrease from 100 to 80 moles and the agriculture ($NH_3$) emissions from 50 to 40 moles. When only transport is reduced, the 80 remaining moles of $NO_2$ react with the 50 moles of $NH_3$ leading to 50 moles of PM. The PM concentrations remain

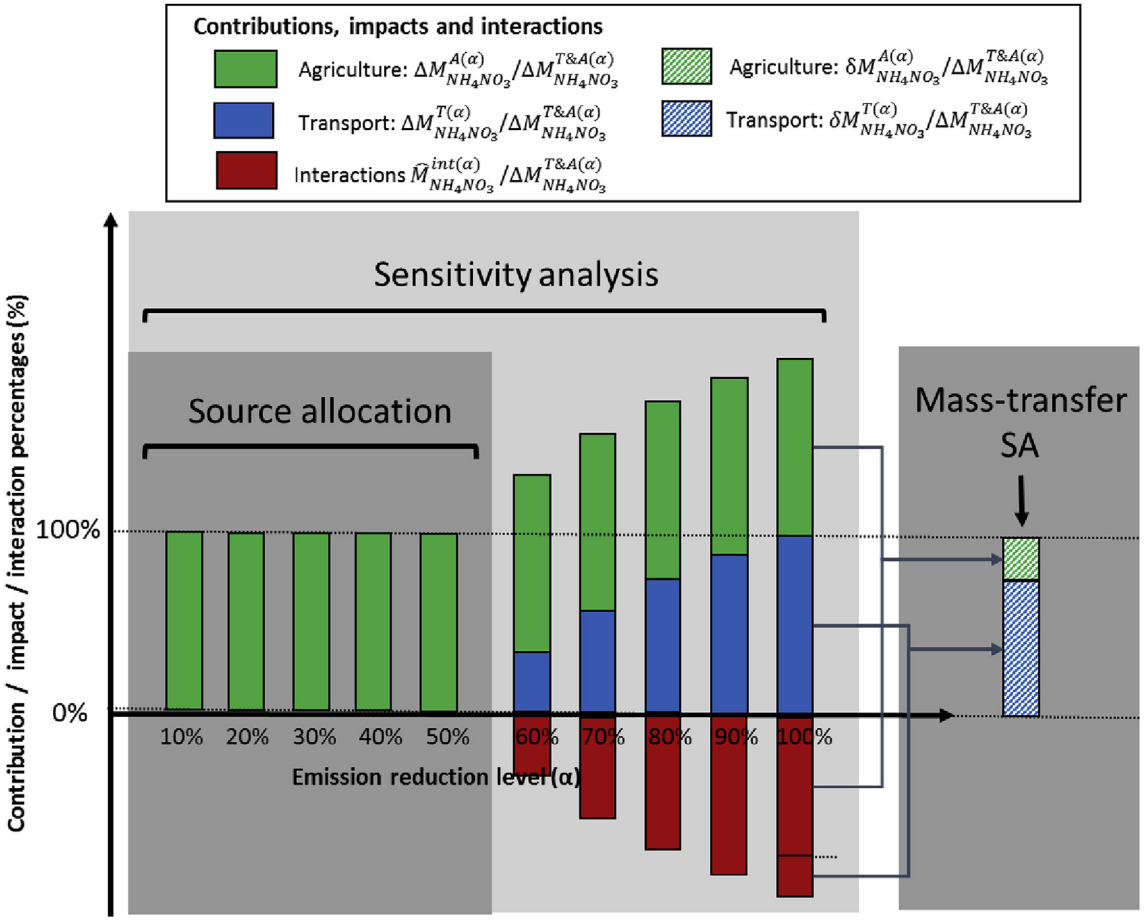


**Fig. 8.** Relative decomposition of the impact of a simultaneous reduction of all precursors ($\Delta M^{T\&A(\alpha)}_{NH_4NO_3}$) using different methodologies for the specific example ($NH_3$ limited regime) presented in the text. (1) Sensitivity analysis is performed for different levels of emission reductions (α) and decomposes $\Delta M^{T\&A(\alpha)}_{NH_4NO_3}$ into different components, the impact of agriculture emissions only (green), the impact of transport emissions only (blue) and the interactions between agriculture and transport emissions (red). (2) Mass-transfer SA decomposes $\Delta M^{T\&A(\alpha)}_{NH_4NO_3}$ into contributions (dashed blue and green), each associated to a given emission precursor. These contributions are obtained from the 100% sensitivity analysis components by combining the direct impacts (green & blue components) with the interaction term (red component), following a precursor mass-ratio apportionment rule. See text for details. (3) Similarly to mass-transfer SA, the source allocation decomposition consists of precursor contributions summing up to $\Delta M^{T\&A(\alpha)}_{NH_4NO_3}$. However, unlike mass-transfer SA, source allocation contributions are equal to the impacts obtained with sensitivity analysis. To keep this property, the interactions terms must be neglected, implying a range of applicability limited to emission reduction levels (α) that are small enough. Source allocation is only valid over the range of emission reductions where no interaction is present. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

therefore unchanged: $\Delta M^{T(20\%)}_{NH_4NO_3} = 0$. When only agriculture is reduced, the 40 moles of $NH_3$ react with the 100 moles of $NO_2$ leading to 40 moles of PM: $\Delta M^{A(20\%)}_{NH_4NO_3} = 50 - 40 = 10$. When transport and agriculture emissions are reduced simultaneously, 80 moles of $NO_2$ react with 40 moles of $NH_3$ leading to 40 moles of PM: $\Delta M^{T\&A(20\%)}_{NH_4NO_3} = 50 - 40 = 10$. The numbers can be used to calculate the interaction term that is equal to zero in this case because only the agriculture emission reduction produces an impact on PM concentrations: $\hat{M}^{int(20\%)}_{NH_4NO_3} = 0$. For emission reductions that are limited in intensity, the chemical regime remains $NO_2$ saturated (or $NH_3$ limited). But a change of regime can occur when more important reductions are considered.

- If α = 80%, transport emissions decrease from 100 to 20 moles and agriculture emissions from 50 to 10 moles leading to $\Delta M^{T(80\%)}_{NH_4NO_3} = 50 - 20 = 30$ , $\Delta M^{A(80\%)}_{NH_4NO_3} = 50 - 10 = 40$, $\Delta M^{T\&A(20\%)}_{NH_4NO_3} = 50 - 10 = 40$ and $\hat{M}^{int(80\%)}_{NH_4NO_3} = 40 - 30 - 40 = -30$. In this situation, the chemical regime is not anymore $NO_2$ saturated and both $NO_2$ and $NH_3$ reductions have an impact on the PM reduction. Fig. 8 shows the sensitivity analysis terms, reported for different levels of emission reductions (α). This figure highlights the interaction terms that progressively gain in importance for the highest levels of emission reductions.

A sensitivity analysis is, a priori, easy to implement using the so called “brute-force” method. For two precursors (as in our example), this method only requires running four air quality model simulations: one base case using the full emissions and three scenarios, one with only transport emissions reduced, one with only agriculture emissions reduced and one with both transport and agriculture emissions reduced. In practice, the “brute-force” method shows quickly its limitations. Indeed, as shown in the previous example, the sensitivity analysis terms depend on the reduction percentage α and three scenario runs would be necessary for each percentage reduction to be analysed. Moreover, the number of scenarios (N) quickly increases with the number of precursors ($n_p$) to be considered ($N = 2^{n_p} - 1$). For three precursors, seven scenario runs would be required for each reduction percentage to estimate all terms. Therefore, the number of scenarios becomes very rapidly prohibitive. In addition, the interpretation becomes challenging due the large number of terms appearing in the analysis. Two approaches can be followed to reduce the complexity related to

"Alpert and Stein" sensitivity analysis: mass-transfer SA and source allocation.

**Mass-transfer SA** decomposes the pollutant mass into a sum of contributions, each associated to a given source. In our example, the mass of PM [$NO_3NH_4$] is split into a mass of $NO_3$ that is attributed to the transport sector (because it originates from the $NO_2$ emissions from this sector) and into a mass of $NH_4$ that is attributed to the agriculture sector (because it originates from the $NH_3$ emissions from this sector). The sectoral decomposition is then simply obtained by applying the molar mass ratio of $NO_3$ and $NH_4$ to the total PM concentration:

$$a^{NO_3} = M_{NO_3}/\left(M_{NO_3} + M_{NH_4}\right) = 62/80 = 0.775 \text{ and, } a^{NH_4} = M_{NH_4}/\left(M_{NO_3} + M_{NH_4}\right) = 18/80 = 0.225 = 1 - a^{NO_3}$$

The 50 initial moles of PM[$NO_3NH_4$] can then be shared between the contribution of the transport, $\delta M^{T}_{NH_4NO_3}$ and the contribution of the agriculture $\delta M^{A}_{NH_4NO_3}$ which are equal to:

$$\delta M^{T}_{NH_4NO_3} = 50 \times a^{NO_3} = 38.75 \text{ and } \delta M^{A}_{NH_4NO_3} = 50 \times a^{NH_4} = 11.25$$

The 50 moles of PM[$NO_3NH_4$] are the result of the simultaneous emissions of 100% of all precursor (transport and agriculture). By definition, the 50 moles of PM are therefore equivalent to the effect of a simultaneous reduction of all precursor emissions (a reduction of all precursor emissions will also decrease the PM by 50 moles). As shown by Clappier et al. (2017), the sum of all contributions can be written as follows:

$$\Delta M^{T\&A(100\%)}_{NH_4NO_3} = \delta M^{T}_{NH_4NO_3} + \delta M^{A}_{NH_4NO_3}$$

A priori, the results of the mass-transfer SA approach are easier to interpret than those of the sensitivity analysis. The number of terms is reduced because no interaction term is visible and it only refers to one reduction percentage (100%). But although not visible, these interaction terms are present. As detailed in Clappier et al. (2017) they are split according to a mass ratio weighting and are accounted for in the precursor contributions (see Fig. 8). Consequently, these contributions are not equal anymore to a "pure" precursor impact as computed with the sensitivity analysis. They do not provide therefore information on the impacts of emission reductions unless the relationship between emissions and concentrations is linear. In our example, mass-transfer SA would estimate a contribution of 38.75 moles for transport while the impacts of an emission reduction in the transport sector would vary between 0 (for reductions up to 50%) to 50 moles (for a 100% reduction) for sensitivity analysis. Under non-linear conditions, this approach cannot be used to estimate the consequence of abatement strategies.

**Source allocation** is produced via CTMs (using "brute-force" methods or decoupled direct method (DDM) methods (e.g. Dunker, 1984; Dunker et al., 2002) or via simplified CTMs (e.g. Source Receptor Relationship like GAINS (Amann et al., 2011) or SHERPA (Thunis et al., 2016). Similarly to mass-transfer SA, source allocation splits the impact of a simultaneous reduction of all precursors into a sum of contributions, each associated to a given emission precursor. However, unlike mass-transfer SA contributions, source allocation contributions are equal to the impact of each precursor reduction computed with the sensitivity analysis. Source allocation contributions are obtained by assuming that a percentage reduction ($\alpha$) can always be found for which the relationship between emission and concentration is linear. The interaction terms can then be neglected and the impact of the reduction of all precursors can be decomposed into the sum of individual precursor contributions:

$$\Delta M^{T\&A(\alpha)}_{NH_4NO_3} \approx \Delta M^{T(\alpha)}_{NH_4NO_3} + \Delta M^{A(\alpha)}_{NH_4NO_3}$$

The main challenge of this approach is to identify the threshold percentage beyond which the linear assumption is no longer valid. In our simple example, the linear assumption remains valid (no interaction terms) for emission reduction that remain below $\alpha = 50\%$, i.e. as long as the chemical regime remains $NO_2$ saturated and the non-linear term is zero (Fig. 8). Thunis and Clappier (2015) showed that for CTM simulations dealing with real situations, a similar level of emission reductions (i.e. up to 50%) remains a valid range for source allocation.

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